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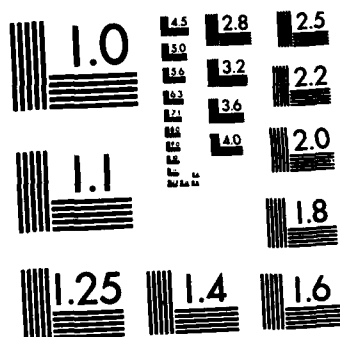UNCLASSIFIED　N00014-80-C-0472　　　　　　　　　　F/G 7/4　　　NL

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of Two Atoms on a Metallic Surface

by

Xi-Yi Huang, K. C. Liu and Thomas F. George

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Department of Chemistry
University of Rochester
Rochester, New York   14627

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SURFACE PLASMONS
PHOTON TRAPPING

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RESONANCE FLUORESCENCE AND PHOTON TRAPPING OF
TWO ATOMS ON A METALLIC SURFACE

XI-YI HUANG, K. C. LIU AND THOMAS F. GEORGE
Department of Chemistry, University of Rochester,
Rochester, New York 14627

ABSTRACT

Surface-dressed optical Bloch equations are derived for
the purpose of evaluating the resonance fluorescence spectrum
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the two-adatom-surface system, and this is studied by means
of a theory which treats the atoms and their surface images
on the same footing.

INTRODUCTION

There has been interest during the past several years in the following
radiation effects at surfaces: (i) the anomalously high intensity of light
inelastically scattered from adsorbed molecules [1] and (ii) the oscillatory
variation of the lifetime of an excited molecule fluorescing near an inter-
face due to interference effects between photon reflection and absorption at
a surface [2]. These phenomena have motivated us to examine the fundamental
processes involved in surface-modified excitation of a laser-driven quantum
system, and for this purpose we have derived a set of surface-dressed optical
Bloch equations to evaluate the resonance fluorescence spectrum of a two-
level atom near or adsorbed on a metallic surface. In this paper, we shall
first review our work on resonance fluorescence of a single adatom [3-5] and
then make an extension to the case of two interacting identical atoms.

The second general topic in this paper is photon trapping. It is well
known that the radiation behavior of an atom in the presence of other
identical atoms is significantly different from that of the independent
atom when the interatomic distance is smaller than the wavelength of the
radiation [6,7]. In this case the atoms are correlated via their inter-
action with the common radiation field and should be treated as a single
quantum-mechanical system with internal degrees of freedom [8]. It is
equally probable for a photon to be emitted or trapped. The emission pro-
cess is known as superradiance, where the intensity is proportional to the
square of the number of atoms [8]. When the atoms are close to a metallic
surface, the photon emitted from one of them can reach another one either
by direct transmission or through reflection from the surface, as illustrated
by FIG. 1. This situation requires that the atoms and their surface images
be considered on the same footing [9,10]. Using this procedure, we shall
consider the spontaneous emission from two adatoms on a metallic surface,
where it will be shown that a photon can be trapped within the two-adatom-
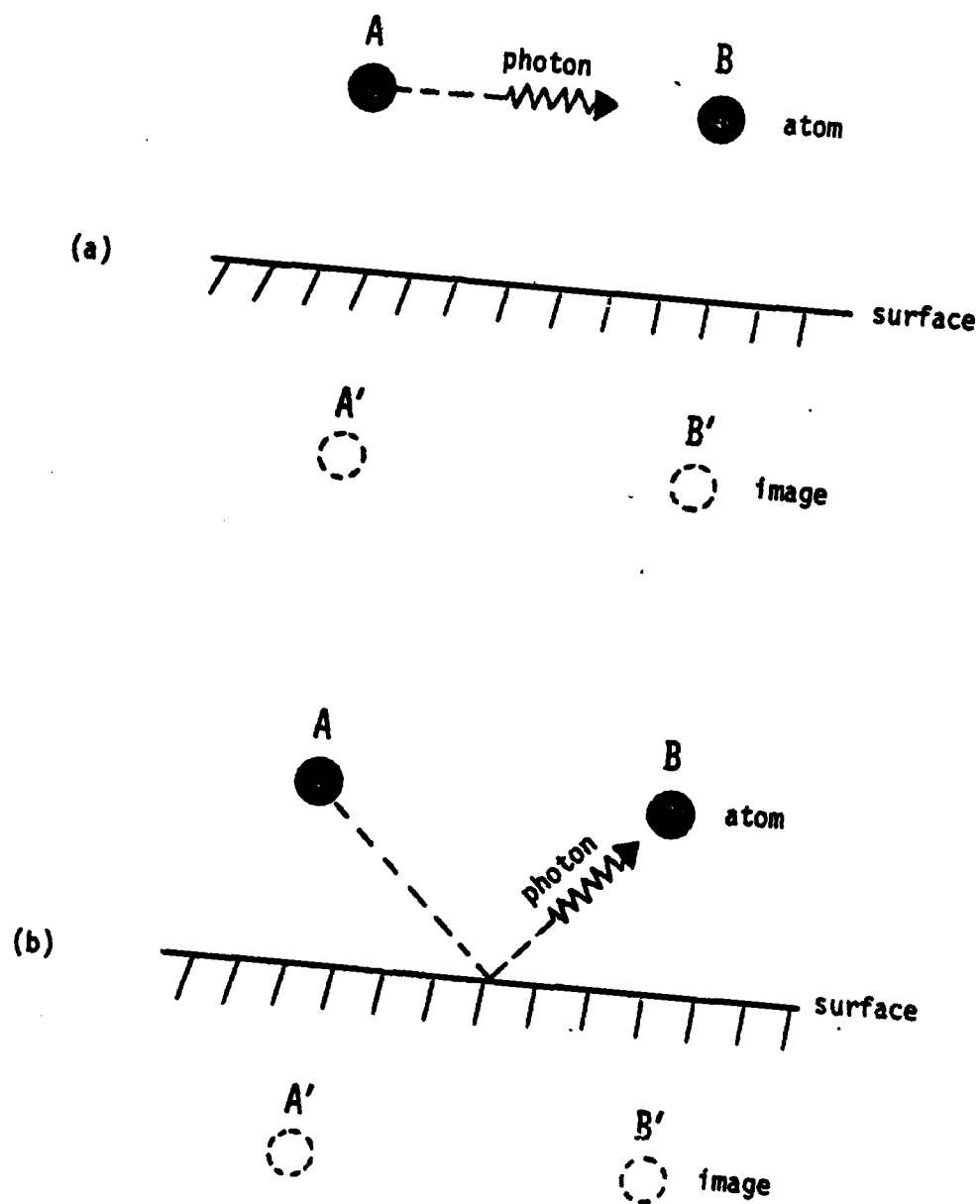surface system under the appropriate circumstance.

FIG. 1. (a) Direct photon exchange
(b) Photon exchange via reflection from the surface

RESONANCE FLUORESCENCE

Resonance fluorescence resulting from the interaction of a two-level system and coherent radiation has been a subject of considerable interest [11-13]. The predicted shapes of the fluorescence spectrum have been verified experimentally [14]. High-resolution experiments have reached such a level of sophistication that higher-order effects such as the contribution of the finite driving laser linewidth [15], the atomic recoil [16] and cooperative atomic interactions [17] can now be analyzed. In a different context, the lifetime of an excited adspecies fluorescing near a gas-metal interface can be influenced substantially by the interference between photon reflection and absorption at the surface. By placing a layer of dye molecules on top of a number of layers of fatty acid which are on a metal surface, it has been possible to observe the fluorescence lifetime of a molecule at a fixed distance from a metal surface. [18].

The above work suggests that it would be interesting to investigate the basic behavior of the surface-modified resonance light scattering spectrum of a laser-driven two-level atom near or adsorbed on a metal surface. The field emitted by the induced atomic dipole is reflected by the surface. The reflected field can be expressed as $\hat{E}_R(t) = \mu_{12}\hat{\sigma}_{12}(t)f(d) + \mu_{21}\hat{\sigma}_{21}(t)f^*(d)$, where d is the distance between the adatom and the surface, μ_{ij} is the electric-dipole transition matrix element between the adatomic states $|i\rangle$ and $|j\rangle$, $\hat{\sigma}_{ij} \equiv |i\rangle\langle j|$ is the adatomic transition operator, and f(d) is a distance-dependent function which has been discussed elsewhere [2-5]. Accounting for the surface-reflected photons and the surface plasmon resonance, we can write the surface-dressed optical Bloch equations (SBE) in the following form (using the rotating-wave approximation and the equilibrium condition $W(0) = -1$, where $W(t) \equiv \hat{\sigma}_{22}(t) - \hat{\sigma}_{11}(t)$ is the population inversion of the adatom):

$$\frac{d}{dt}\begin{pmatrix} \hat{S}_{21}(t) \\ \hat{W}(t) \\ \hat{S}_{12}(t) \end{pmatrix} = \begin{pmatrix} -\tilde{\gamma}_2+i\Delta & i\Omega^-(t)/2 & 0 \\ i\Omega^+(t) & -\gamma_1 & -i\Omega^-(t) \\ 0 & -i\Omega^+(t)/2 & -\tilde{\gamma}_2-i\Delta \end{pmatrix}\begin{pmatrix} \hat{S}_{21}(t) \\ \hat{W}(t) \\ \hat{S}_{12}(t) \end{pmatrix} - \gamma_1\begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}. \quad (1)$$

The phase-fluctuating Rabi frequency is $\Omega^\pm(t) \equiv \Omega\exp[\mp i\phi(t)]$, where $\Omega \equiv (2/\hbar)|\mu_{12}|E_0(t)$ and E(t) is written as an expectation value in a coherent state of the laser field in terms of the phase factor $\phi(t)$ and the laser frequency ω_L as $E(t) = E_0(t)\exp[-\omega_L t + i\phi(t)]$; $\Delta \equiv \omega_{21} - \omega_L$ is the detuning, where ω_{21} is the adatomic transition frequency; γ_1 is the population relaxation constant; and $\tilde{\gamma}_2 \equiv \gamma_2 + \gamma_\Delta$, where γ_2 is the surface-free phase relaxation constant and $\gamma_\Delta \equiv (2/\hbar)\text{Im}(f)|\mu_{12}|^2$ is the surface-induced phase relaxation constant which has been evaluated by means of the Drude model [19] for the metal medium, whereby the dielectric constant is expressed simply in terms of the emission frequency, the inverse relaxation time and the plasma frequency.

In the case of a weak driving field or large detuning, we can set W(t) approximately equal to -1, i.e., the atom almost stays in its ground state. The effects of the laser bandwidth γ_L can be included by means of the phase-diffusion model [20,21], $\langle\langle\Omega^-(t_1)\Omega^+(t_2)\rangle\rangle = \Omega^2\exp(-\gamma_L|t_2-t_1|)$, where the double bracket signifies an average over the stochastic ensemble and a standard quantum mechanical average. The power spectrum $S(\omega)$ of scattered light, given by the Fourier transform of the dipole-dipole correlation function $\langle\langle\hat{S}_{21}(t_2)\hat{S}_{12}(t_1)\rangle\rangle$, can be obtained analytically from the SBE [4]. This weak-field spectrum exhibits two peaks: the Rayleigh scattering peak at $\omega = \omega_L$ and the fluorescence peak at $\omega = \omega_{21}$. The shape of the former is determined mainly by γ_L, while the latter is strongly dependent on the adatom-surface distance, such that the peak is raised and broadened as the adatom is brought closer to the surface.

In the case of a strong driving field, $\Omega^2 + \Delta^2 \gg \tilde{\gamma}^2$, if we set γ_L equal to zero we have again been able to obtain an analytic expression for the power spectrum $S(\omega)$, which now exhibits three peaks, in contrast to the two-peak spectrum of the weak-field case [5]. The central peak at $\omega = \omega_L$ corresponds to Rayleigh scattering. The lowest frequency peak at $\omega = \omega_L - (\Delta+\delta)$, where $\delta = \Delta\{[1+(\Omega/\Delta)^2]^{1/2} - 1\}$, corresponds to a three-photon process (absorption of two laser photons and spontaneous emission of a photon). The highest frequency peak at $\omega = \omega_L + (\Delta+\delta)$ is called the flourescence component of the spectrum. When the adatom-surface distance is very large, the (surface-free) spectrum is symmetric, where the two side peaks have the same height (intensity) and are lower than the central Rayleigh peak. When the adatom-surface becomes smaller, the symmetry is destroyed due to the interference between the incident laser field and the surface-reflected field.

The comparison with the pure gas-phase situation for the case of positive detuning is interesting, where the random collision interruption always causes the fluorescence peak to be higher than the three-photon peak. For the surface-modified process this does not always occur, since there is an oscillatory distance dependence of the emission intensity due to the interference mentioned above. While the fluorescence component is generally larger, the three-photon component is larger for some distances. This is a unique behavior at a surface which has not been predicted for the usual gas-phase situation [22].

We now extend our study to resonance fluorescence in a system of two interacting two-level atoms, which has previously been analyzed for the gas-phase situation [23-25] where a special interest is in the possible observation of an optical bistability in a many-atom system. While we have briefly considered the resonance fluorescence of many interacting atoms near a metal [26], here we want to concentrate on the particular case of two identical two-level atoms near or adsorbed on a metal surface. We assume the distance R between the two adatoms to be sufficiently small, i.e., $R \leq 2\pi/k$ where k is the laser wavenumber, so that we can adopt the global dipole approximation which eliminates the differences of the electromagnetic field exerted on the two atoms. However, we must include the dipole-dipole interaction, derived from the first-order dispersion forces, between the adatoms.

The resulting SBE take the following form as an extension of Eq. (1), where $i\neq j=1,2$ label the two adatoms and h.c. stands for Hermitian conjugate:

$$(\dot{\hat{\sigma}}_{12})_i = -(i\omega_{21}+\tilde{\gamma}_2)(\sigma_{12})_i - \frac{i}{2}\Omega\hat{W}_i + \Gamma_{12}\hat{W}_i(\hat{\sigma}_{12})_j$$

$$- \frac{2i}{\hbar}\,\text{Re}[f(d)]|\mu_{21}|^2\hat{W}_i(\hat{\sigma}_{12})_j - \frac{2i}{\hbar}\,f^*(d)\mu_{21}^2\hat{W}_i(\hat{\sigma}_{21})_j \qquad (2)$$

$$\dot{\hat{W}}_i = -\gamma_1(1+\hat{W}_i) - [i\Omega(\hat{\sigma}_{21})_i + 2\Gamma_{12}(\hat{\sigma}_{12})_i(\hat{\sigma}_{12})_j + \text{h.c.}]$$

$$+ \{i\,\frac{2f(d)}{\hbar}\,|\mu_{21}|^2[(\hat{\sigma}_{21})_i(\hat{\sigma}_{12})_j - (\hat{\sigma}_{21})_j(\hat{\sigma}_{12})_i]$$

$$- i\,\frac{2f(d)}{\hbar}\mu_{12}^2[(\hat{\sigma}_{12})_i(\hat{\sigma}_{12})_j + (\hat{\sigma}_{12})_j(\hat{\sigma}_{12})_k] - \text{h.c.}]. \qquad (3)$$

We have assumed a fully coherent laser field, such that $\gamma_L = 0$ and the Rabi frequency is independent of time. The adatom dependence of this frequency has been omitted as a consequence of the global dipole approximation. The terms containing Γ_{12} account for the mutual interaction of the two adatoms through their near-dipole fields and exchange of photons, where the

R-dependent quantity Γ_{12} is the sum of the R-dependent damping $1/\tau_{12}$ and the frequency shift Ω_{12} [6].

The power spectrum of the scattered light is given by the Fourier transform of the sum of correlation functions $g_i^* g_j < [\hat{\sigma}_{21}(t)]_i [\hat{\sigma}_{12}(t+\tau)]_j >$ over i and j, where g_i is the geometric factor and the limit $t \to \infty$ is taken. Some interesting features of the spectrum are the following: (i) The three peaks of the strong-field case are further split due to the adatom-adatom interaction. (ii) This interaction and the surface-reflected field lead to an asymmetric spectrum, as discussed earlier in this paper for the single-adatom case and in Ref. 27 for the surface-free case. (iii) Analogous to the single-adatom case, the peak heights depend strongly on the adatom-surface distance and resonance condition of the surface palsmons, where such dependence is oscillatory due to the interference between the driving and reflected fields.

PHOTON TRAPPING

We consider two adatoms A and B near a metal surface (see FIG. 1), where the interactomic and adatom-surface distance are smaller than the radiation wavelength, where the radiation corresponds to emission from A in an excited state (we assume the intital excitation of A to be independent of subsequent emission and absorption). The states of the adatoms and their associated surface images can be expressed as [10]

$$|\psi_\pm>_X = \frac{1}{\sqrt{2}} \{ |\alpha>_X|\beta>_{X'} \pm |\beta>_X|\alpha>_{X'} \},\qquad(4)$$

where the subscript X is either A or B, the prime signifies the image, $|\alpha>$ and $|\beta>$ are the excited and ground adatomic states, respectively, and the + and - signs correspond to the adatomic transition dipole oriented perpendicular and parallel to the surface, respectively. We shall neglect the diploe-dipole interaction between the two adatoms, since here we are not interested per se in the frequency shift of the emitted photon [28]. Hence the interaction Hamiltonian for the two adatoms is simply $H_{int} = H_A + H_B$, where $H_A (H_B)$ is the interaction Hamiltonian for A(B). Within the dipole approximation, the diagonal matrix elements of H_A and H_B are zero, i.e., $_X<\alpha|H_X|\alpha>_X = {}_X<\beta|H_X|\beta>_X = 0$, and the off-diagonal elements equal the same finite amount Δ, i.e., $_X<\alpha|H_X|\beta>_X = {}_X<\beta|H_X|\alpha>_X = \Delta$.

We shall consider two cases for different dipole orientations. In the first case, both dipoles are perpendicular to the surface, whereby the state of the total system can be written as

$$|\chi_\pm> = \frac{1}{\sqrt{2}} \{ |\psi_+>_A|g>_B \pm |g>_A|\psi_+>_B \},\qquad(5)$$

where $|g>_X = |\alpha>_X|\alpha>_{X'}$ represents the ground state of adatom X. The ground state of the total system is $|G> = |g>_A|g>_B$, and it is easy to verify that the state $|\chi_->$ has a vanishing decay rate since $<G|H_{int}|\chi_-> = 0$. Thus, if the system is in the state $|\chi_->$, the photon will be trapped. In the second case, both dipoles are parrallel to the surface, whereby the state of the total system is

$$|\chi_\pm> = \frac{1}{\sqrt{2}} \{ |\psi_->_A|g>_B \pm |g>_A|\psi_->_B \}.\qquad(6)$$

Again, it can be shown that the state $|\chi_->$ leads to photon trapping, i.e., $<G|H_{int}|\chi_-> = 0$.

SUMMARY

Surface-dressed optical Bloch equations have been solved for the resonance fluorescence spectrum of a two-level atom near or adsorbed on a metal surface, taking into account the influence of reflected photons, dephasing due to atomic collisions, the linewidth of the driving laser field and the resonance excitation of surface plasmons. In the weak-field or large-detuning limit, the spectrum exhibits two peaks, one corresponding to Raleigh scattering and the other to fluorescence. In the case of a strong field, there are three peaks: the central peak corresponding to Raleigh scattering, the lowest frequency peak corresponding to a three-photon process and the highest frequency peak called the fluorescence component of the spectrum. The surface-free symmetry of the spectrum is destroyed for finite adatom-surface distances due to the interference between the incident laser field and the surface-reflected field. The nature of the asymmetry is different than that induced by collisions in a pure gas-phase system. The spectrum for a system of two interacting two-level atoms is further split due to the adatom-adatom interaction. A photon emitted by an excited adatom can become trapped by the two-adatom-surface system, and this phenomenon has been examined in terms of adatomic states and their surface images.

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